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Numerous epidemiological studies have demonstrated associations between ambient combustion-source particulates and adverse health outcomes. In order to better understand exposure to particles, we evaluated a portable particle counter for its ability to measure short-term peaks in personal particle exposure associated with various activities, such as proximity to vehicular traffic. In a series of laboratory and field measurements, a hand-held particle counter was evaluated by collecting simultaneous filter samples of particulate matter less than 2.5 µm (PM.s) using a personal monitor. Time activity information was collected using a Personal Digital Assistant (PDA) which allows for linking of exposure events and particle measurements with 1 min temporal resolution, Laboratory and field experiments comparing the particle counter with the personal PM_{2.5} samples indicated low correlations ($R^2 < 0.39$) for all size ranges. Despite these rather poor correlations, field measurements collected during different commuting modes (walking/cycling, car, bus, subway) and in different microenvironments indicated the ability of the particle counter to measure short-term particle exposures, especially those associated with combustion sources. Stratifying the measured particle counts by proximity to different particle sources enabled us to identify activities/microenvironments which were associated with higher exposures. Outdoor particle counts were significantly higher than indoor counts for particles smaller than 5.0 µm. Significantly elevated particle exposures were associated with proximity to environmental tobacco smoke (ETS), cooking emissions, wood smoke and with travel in vehicles powered with internal combustion engines.

Keywords: mobile sources, particles, particle counts, personal digital assistant, personal exposure,

Introduction

Studies assessing inhalable (PM₁₀) and fine (PM_{2.5}) particle exposure and health outcomes have linked ambient particulate levels to increases in mortality, respiratory symptoms, health service utilization, work or school absenteeism, and to decreases in lung function (Dockery and Pope, 1994; Dockery et al., 1993; Pope et al., 1995). While adverse health outcomes have been linked to increases in 24-h average ambient particulate concentrations, little is known about shorter duration exposures and their impact on human health. To broaden our understanding of this relationship, transient exposures and the instruments used to measure them need to be evaluated. In particular, outdoor exposure to mobile source emissions and exposures encountered while in transit have not been evaluated in most previous particle exposure studies, despite evidence suggesting the importance of characterizing these exposures to reduce misclassification in epidemiological studies.

Vehicle emissions are known as a major source of PM2 s in most urban areas (Wilson and Suh. 1997). Source apportionment studies in the Vancouver, British Columbia area, where this study was conducted, suggest that 35-45% of PM24 mass originates from direct motor vehicle emissions (Lowenthal et al., 1994). Increased respiratory symptoms in children are associated with living near a freeway and with traffic density, especially truck traffic (Van Vliet et al., 1997). An assessment of particulate matter near urban roadways (Balogh et al., 1994) has shown that direct tailpipe emissions, especially diesel vehicle emissions, are more important contributors to mobile source PM2.5 emissions than resuspension of settled particulate. A recent study of spatial variability in particulate concentrations has shown that PM2.5 concentrations near major roads were 30% greater than at a background location not influenced by local traffic (Janssen et al., 1997). Black Smoke levels were 2.6 times higher at the roadside locations, indicating the important contribution of diesel exhaust to traffic-related PM25 emissions.

The objectives of this study were: (a) to evaluate the ability of a continuous light-scattering particle counter to detect short-term variation in particle counts as assessed by time-activity measurement of proximity to particle sources, (b) to compare exposures assessed using the particle counter to those determined using a personal PM2.5 impactor, (c) to evaluate a Personal Digital Assistant (PDA) for shortduration time-activity data collection, and (d) to conduct a

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^{1.} Abbreviations: ETS, environmental tobacco smoke; PDA, Personal Digital Assistant; PEM, personal environmental monitor; PM2.5, particulate matter less than 2.5 μ m; PTEAM, Particle Total Exposure Assessment Methodology.

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preliminary assessment of continuous personal particle exposure. We also used the particle counter to evaluate the personal cloud phenomenon. The personal cloud effect was described in detail in manuscripts reporting on the US Environmental Protection Agency's Particle Total Exposure Assessment Methodology (PTEAM) study. In this study, daytime personal PM₁₀ exposures were more than 50% higher than either outdoor or indoor levels. It was suggested that this observation was a result of resuspension of coarse particles by human activity or due to the proximity of individuals to particle sources (Wallace, 1996).

Commuting modes were measured in the Vancouver metropolitan area. These included driving, walking, cycling, and public transit. Commuting by public transit included three modes: Skytrain—an electric-powered rapid transit train, electric trolley or diesel-powered buses, and Seabus—a diesel-powered hovercraft. To assign particle counts as a function of exposure variables, field measurements were made during ten experiments, each with scripted activity patterns.

To evaluate short-duration particle exposures, we used a hand-held PDA, also known as a hand-held pen computer, to record, in real time, certain activities and exposure microenvironments and to assign them to measured exposure peaks. Similar PDA devices have been used in occupational exposure assessment to record information on job-tasks or exposure surveys (Haskew et al., 1995; Cohen and Cotey, 1997; Wilkins et al., 1997). We were interested in the suitability of such devices for use as real-time activity logs in exposure assessment studies.

Methods

Continuous personal particle exposure was assessed using a light scattering airborne particle counter (APC-1000, Biotest Diagnostics). This small, quiet, battery-operated instrument measures particle number concentrations (particles per cubic centimeter) in four different size ranges: ≥ 0.3 -<0.5 μ m, ≥ 0.5 -<1.0 μ m, ≥ 1.0 -<5.0 μ m and ≥ 5.0 μ m. Air is drawn into the instrument at a 2.8 ν min flow rate. Particles are sized and counted by scattering of 820 nm light

Table 1. Activity sequence for field experiments.

Sequence	Activity	Duration (min) 30			
1	Stationary outdoors				
2	Transit	Variable (30-56)			
3	Stationary outdoors	30			
4	Indoors	Variable (25-200)			
5	Stationary outdoors	30			
6	Transit (same mode as 2)	Variable (30-45)			
7	Stationary outdoors	30			

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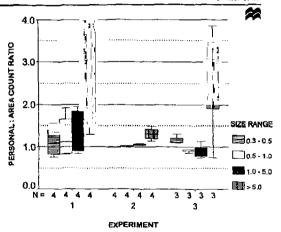


Figure 1. The effect of particle size on the 'personal cloud.' *Experiment 1 involved a subject who was mobile within a small dormitory room. The subject remained seated during Experiments 2 and 3. N refers to the number of comparisons in each experiment.

emitted from a laser diode. Two-minute average concentrations were logged over 6-8 h measurement periods. In addition to particle concentrations, the particle counter also recorded temperature and relative humidity. The particle counter was factory-calibrated before and after the study was conducted using polystyrene nanospheres in accordance with ASTM Standard F649.

Fine particles were collected using a personal PM2 5 impactor (Personal environmental monitor, PEM: MSP Corp.) connected to a 6-in, long aluminum inlet. Samples were collected at a 4 l/min flow rate with a personal sampling pump (SKC Inc., PCXR4). Flows were measured before and after the sampling period with a precision rotameter (Matheson 603) calibrated with a frictionless piston (BIOS Corp.). Particles were collected on 37 mm Teflon filters (Gelman, R2PJO37) which had been preweighed in triplicate using a micro-balance (Sartorius M3P) (1 μ g resolution, $\pm 2 \mu$ g sensitivity) after equilibration for 48 h in a temperature/humidity controlled weighing room (25±0.5°C, 43±5% relative humidity). After sample collection, these same filters were reweighed, again in triplicate, using the same micro-balance and protocol. Filter masses were lab blank corrected. The mean (absolute change) change in lab blanks was 5 μg (range ±14 μg), while the mean change in filter mass for all samples was 46 μ g. The detection limit, based on three times the standard deviation of blank filters and assuming a 6-h sample duration, was 10.4 μ g/m³. This is only slightly higher than the 8.3 μ g/m³ reported detection limit for 12-h samples using the same sampler as described in the PTEAM study. As part of the PTEAM study, precision experiments conducted with the PM₁₀ version of this impactor indicated a mean relative

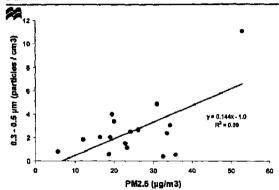


Figure 2. Relationship between PEM PM_{2.5} mass concentrations and particle counts for 17 field experiments, including the ten personal sampling measurements and seven experiments with the particle counter and PEM co-located either indoors or outdoors.

standard deviation of 6% for collocated pairs of personal samplers worn by subjects (Thomas et al., 1993). We conducted three colocation experiments and found relative standard deviations of 4, 9 and 13% (mean=9%) for mass concentrations of 5-34 µg/m³.

Laboratory Experiments

Laboratory studies were conducted to evaluate the personal cloud effect. For these experiments, the particle counter was worn by a subject for 15 min and then placed 1-3 m away on a desk for 15 min, before repeating this sequence for the duration (approximately 2 h) of the experiment. During the first experiment, the subject walked about a small dormitory room, while he remained seated at a laboratory desk during the second and third experiments.

Field Experiments

Ten 6-h field experiments involving scripted activity patterns were conducted during May-October. During these experiments, personal particulate exposures were measured using the particle counter worn next to a PM2.5 PEM. The activity pattern was designed to simulate morning and afternoon commutes to/from work/school, but condensed into a 6-h period by reducing the amount of time spent at the indoor destination. The general sequence followed for all experiments is indicated in Table 1. In the course of these experiments, the following activities and microenvironments were evaluated: indoors with cooking and/or environmental tobacco smoke (ETS) present/absent in the vicinity of the subject, outdoors with ETS present/absent: and during transportation by car, bus, bicycle, Skytrain, Seabus, or on foot. Time-activity information was recorded throughout each experiment whenever an activity/microenvironment changed.

Time-activity logs were created using a PDA (US Robotics/3Com PalmPilot®) using Pilotforms® software (Pendragon Software). The electronic logging form in-

Table 2. Geometric means (GeoMean) (particles/cm³) and standard deviations (GSD) of particle count concentrations for activities/microenvironments, by particle size.

Activity	N .	Particle size range								
		0.3-0.5		0.5-1.0		1.0-5.0		>5.0		
		GeoMean	GSD	GeoMean	GSD	GeoMean	GSD	GeoMean	GSD	
No cooking	1602	1.8	2.1	4.0*	2.2	0.5ª	2.3	0.04ª	2.8	
Cooking	175	1,9	9.1	4.9	2.3	1.0	1.8	0.07	1.7	
No ETS	1756	1.8"	2.1	4.6*	2.2	0.6°	2.3	0.05*	2.7	
ETS	36	4.5	1.4	11.9	1.5	1.1	1.5	0.06	1.9	
Outdoor	347	2.4	2.1	4.9ª	2.0	0.7 ⁸	1.6	0.05	2.3	
Indoor	601	1.5	1.8	3.1	1.8	0.5	2.4	0.04	2.7	
Walking	392	2.6	2.0	5.4	2.0	0.7	1.7	0.95	2.2	
Bicycle	61	2.1	1.5	5.2	1.4	0.7	1.3	0.07 ^b	1.9	
Skytrain	27	2,1	1.6	5.6	1.7	0.7	1.7	0.03 ^b	2.1	
Bus	153	2.2 ^b	1.6	6.4	1.7	1.1 ^b	2.0	0.07 ^b	2.3	
Car	154	3.3 ^b	1.7	6.7 ^b	1.4	0.8	1.8	0.05	1.9	
Seabus	17	5.0 ^b	1.8	12.2 ^b	2.1	1.3 ^b	2,1	0.06	2.1	

^{*} P<0.05.

Indoor measurements include only those without cooking or ETS. Outdoor measurements include only stationary (non-commuting) outdoor measurements. N refers to the number of 2-min average particle count measurements associated with each activity/microenvironment,

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^b Significantly different than walking, p<0.05.

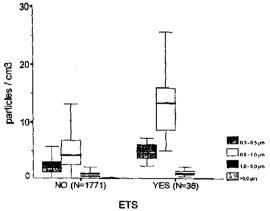


Figure 3. The impact of proximity to ETS on measured personal particle counts for all four size ranges.

cluded fields for indoors (ves/no), outdoors (ves/no)

commuting (car, bus, walk, cycle, Skytrain, other), density

Pilotforms(m) output in Excel(m) spreadsheets. In this way. each 2-min particle count measurement had a series of timeactivity variables attached to it. Statistical analyses of these combined records were conducted with SPSSm software. Particle count data approximated a log normal distribution. Accordingly, transformed data were used for all statistical tests. Both geometric and arithmetic means are reported. Particle concentrations were assessed for different exposures/microenvironments by pooling the ten field experiments, which accounted for a total of 60 h of exposure data.

Results

Laboratory Experiments

Laboratory assessments of the personal cloud using the particle counter showed that the ratio between personal and

of traffic (scale from 1-4), proximity to cooking (yes/no) and proximity to ETS (yes/no). The electronic forms also automatically dated and time stamped all entries. The presence of other suspected particle sources, such as wood

smoke, was also recorded. Particle count data were linked to

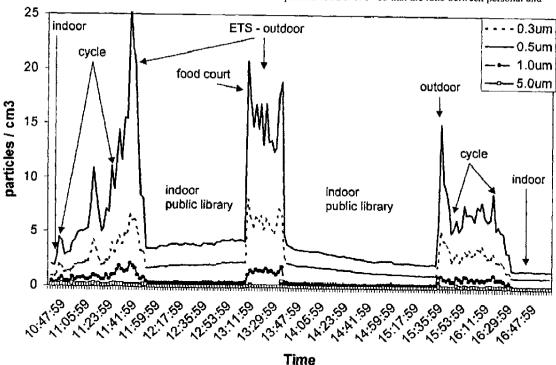


Figure 4. Particle counts in four size ranges measured during a 6-h commuting experiment. Arrows connected at stem indicates entire interval associated with indicated activity/microenvironment.

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area particle counts was greatest for the larger particles. especially those greater than 5.0 µm in diameter (Figure 1). Although these particles represent only a small percentage of the total number of particles, they account for a much larger proportion of the particle mass. Further, the mean personal; area ratio was greater for Experiment 1, which included a modest level of personal activity, than for Experiments 2 and 3, in which the subject was sedentary. To improve our understanding of the relationship between the particle count and mass measurements, we estimated a particle density of 2.8 g/cm³ for Vancouver ambient air particles (this is based on electron microscopy and EDX sizing and determination of particles retained in lungs of Vancouver residents) (Churg and Brauer, 1997). Assuming spherical shape, the 5, 1.0, 0.5 and 0.3 μ m particle size ranges, as counted by the particle counter, would correspond to aerodynamic diameters (d_a) of 3.7, 1.7, 1.2 and 0.9 μ m, respectively. By the same estimate, a 2.5 μ m d_a particle (collected by the PEM) corresponds to an actual size (as measured by the particle counter) of 2.2 μ m. This estimate suggests that particles in the two smallest size ranges measured by the particle counter are also collected by the PEM as are a portion of the particles in the next largest size range. Applying this estimate to our results then implies that the personal cloud effect, while greater for larger particles $(d_a>3.7 \mu\text{m})$, can still be observed for fine particles $(d_a<3.7 \mu\text{m})$, especially when subjects are more active.

Field Experiments

Particle counts in the 0.3–0.5 μ m and 0.5–1.0 μ m ranges were found to be poorly correlated with PM_{2.5} PEM concentrations (N=17, R^2 =0.39 and 0.36, respectively) (Figure 2). Correlations were even lower for the other size intervals; R^2 =0.20 and 0.002 for 1.0–5.0 and >5.0 μ m size ranges, respectively. As discussed above, we estimated the particle mass from the particle count data, by assuming a density of 2.8 g/cm³ for these particles. Using this density and assuming spherical particles, the mass corresponding to

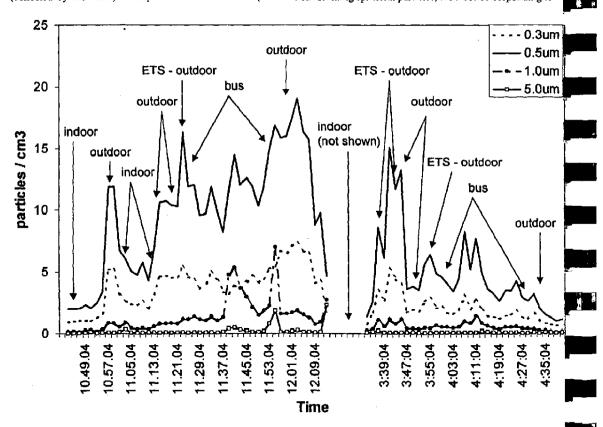


Figure 5. Particle counts in four size ranges measured during a 6-h commuting experiment. A 1.5-h commuting sequence in the morning and a 1-h afternoon commuting sequence are shown. Arrows connected at stem indicates entire interval associated with indicated activity/microenvironment.

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the particle counts was estimated. These estimated mass concentrations were not correlated with particle mass as measured by the PEMs and analyzed gravimetrically.

When considering all field measurements, 28% of particles by number were in the 0.3–0.5 μ m size range and 61%, 10% and 1% in the 0.5–1.0, 1.0–5.0 and >5.0 ranges, respectively. These same percentages were generally consistent for the different microenvironments/activities which we assessed. Across all field experiments, particle counts were highly variable. The relative standard deviation of the particle counts increased with increasing particle size and ranged from 75% to 322%.

Non-commute Activities and Microenvironments Table 2 presents particle counts for the different microenvironments and activities. Exposures can be computed by multiplying the mean particle concentration for each activity/microenvironment by the length of the measurement period. Comparison of indoor measurements (no ETS, no cooking) with outdoor (non-commute) geometric means indicated

that indoor particle concentrations (ANOVA, p<0.0001) were lower than outdoors, for all size intervals. Except for the largest size interval, which measured very few particles (1%), the ratio of indoor to outdoor geometric mean particle counts was approximately 0.62. Cooking was found to significantly elevate indoor particle counts for all size intervals (p<0.05). Visual observation of wood smoke by the subject wearing the particle monitors was associated with significantly elevated outdoor particle counts in the 0.3–0.5 and 0.5–1.0 μ m intervals (p<0.05). Similarly, the observation of ETS either indoors or outdoors was associated with a significant elevation of particle counts in all size intervals (p<0.05) (Figure 3).

Commute Modes The importance (relative to walking) of each of the transportation modes to particle exposures for each of the four particle size ranges is presented in Table 2. Rank orders were mostly consistent for particle diameters less than 5.0 μ m. Commute by combustion-powered vehicles generally was associated with higher particle

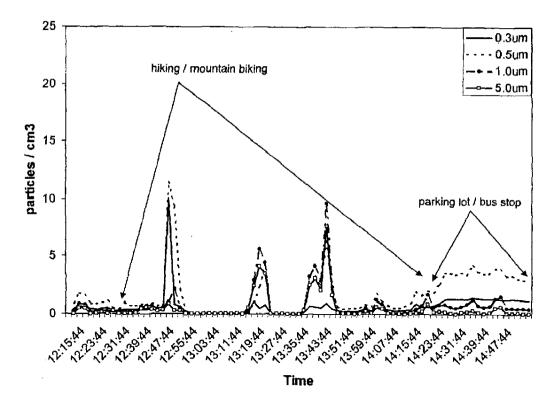


Figure 6. A 2.5-h sequence of particle counts in four size ranges measured during a 6-h experiment. Arrows connected at stem indicates entire interval associated with indicated activity/microenvironment.

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concentrations. Transportation by walking, by bicycle, or on the electric-nowered Skytrain resulted in lower concentrations. This order was different for particles larger than 5.0 um. In this size range, cycling was associated with the highest concentrations, significantly higher than either walking or commuting by car. Transportation by Seabus resulted in significantly higher concentrations than all other transportation modes for measured particles less than 5.0 um, although these measurements may reflect a small sample size. Particle concentrations measured during bus travel were similar to those measured during commuting by car. Transportation by Skytrain was associated with lower concentrations of particles greater than 5.0 µm than all other modes of transportation except the automobile. Our subjective four-point traffic level scale was not useful in predicting particle counts as there was no consistency in the relationship between traffic scale and measured particle

Example temporal plots of particle counts are presented in Figures 4-6. Figure 4 shows a full experiment including particle counts measured indoors, while cycling, and during outdoor ETS exposure over a 6-h period. The highest particle concentrations across all size ranges were measured during the period of outdoor ETS exposure, while the lowest concentrations were measured indoors. This example shows the clear difference between indoor and outdoor levels as well as the more variable nature of the outdoor exposure profile. The relative impact of ETS and cooking as sources of particle exposure is also apparent in this plot. Figure 5 depicts particle counts representing two bus travel periods on the same day. Particle count peaks are evident due to ETS exposure just prior to boarding the bus and after disembarking from the bus, presumably associated with the diesel exhaust of the bus itself. During this 6-h measurement, the highest particle counts for all size ranges were measured outdoors after disembarking from the bus, while the lowest counts were measured indoors. After disembarking from the bus, elevated coarse particle counts are also observed, suggesting exposure from resuspended road dust. Figure 6 shows 45 min of outdoor particle counts, including a period of hiking and cycling along a dirt trail where the influence of coarse particles is evident and the ratio between the particle sizes is different than those indicated in Figures 4 and 5.

Discussion

Laboratory Experiments

For the three laboratory experiments, the mean ratio of personal: area counts were 0.90-1.4 for the three smallest size ranges measured by the particle counter. This is somewhat lower than the personal: area concentration ratios of 1.5-2 measured in the PTEAM study (Wallace, 1996),

although the PTEAM samples were collected from active subjects. Our observation that personal:area ratios (mean ratios of 1.3–3.0) were greatest for the largest particle size range (>5 μ m, which we estimate to correspond to d_a >3.7 μ m) supports the suggestion of Wallace that the personal cloud may be related to resuspension of larger particles (Wallace, 1996). The resuspension theory is also supported by our observation of a higher personal:area particle count ratio for the experiment which included a modest level of personal activity, than for those in which the subject was sedentary.

Field Experiments

The hand-held particle counter, combined with the PDA proved to be a useful tool for following real time changes in particle counts for each of the four size ranges. The instrument performed well with a diverse set of particle sources: cooking, wood smoke, ETS, vehicle exhaust, and resuspended particulate. The simultaneous measurement of particle counts in four size intervals allowed us to differentiate the impact of combustion and resuspension sources in the exposure assessment. Such information is typically not available from personal monitoring studies involving gravimetric particle measurements. For example, in Figures 4-5, particle count peaks associated with combustion sources are primarily restricted to the 0.3-0.5 and 0.5-0.1 µm size intervals, whereas Figure 6 depicts a period of hiking and cycling in which particle counts in the three largest size ranges were elevated.

The low correlations between particle counts in the 0.3–0.5 or 0.5–1 μm range and PEM PM_{2.5} concentrations ($R^2 \le 0.39$) may well reflect differences in the particle size ranges being measured or the variable performance of the particle counter for the different size ranges. This is supported by decreases in the degree of correlation as size intervals increase. Discrepancies between concentrations measured as particle counts and concentrations measured as particle mass may also result from differences in particle density for the different size ranges and for different microenvironments. Our attempts to estimate the particle mass concentrations from the count data produced even lower correlations between the two methods.

While the differential response of different optical particle counting devices to aerosols of different size, shape and density make it difficult to determine the accuracy of the particle counts, the levels measured by the hand-held particle counter in this study were similar to those reported for other microenvironmental measurements using different particle counting instruments. Roshanel and Braaten (1996) used a Climet CI-7000 counter to measure particle counts in the 0.3-5 µm ranges averaged approximately 1.8 particles/cm³ (Roshanel and Braaten, 1996). Leese et al. conducted year-long measurements inside an occupied four-story

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office building using a Met-One 217 optical particle counter. Mean particle counts $(0.5-15~\mu\text{m})$ in different areas of the building were 1.1-1.5 particles/cm³, while the average outdoor level was 4.6 particles/cm³ (Leese et al., 1997). Using the same model particle counter, Weschler and colleagues measured particles for more than 1 year inside an occupied telecommunications office (no smoking and without combustion appliances) in Southern California, Typical outdoor $(0.5-1.0~\mu\text{m})$ particle counts were approximately 8 particles/cm³, but reached as high as 53 particles/cm³. Typical indoor concentrations were 1 particle/cm³ or lower, but occasionally reached levels as high as 35 particles/cm³ (Weschler et al., 1996).

In our study, the highest concentrations of particles less than 5.0 µm in size were measured during travel on the diesel-powered hovercraft called the Seabus. Car and bus travel were the next most important commuting modes in terms of particle exposure. Of the 1053 vehicle fleet serving the greater Vancouver area, 784 buses are powered by diesel engines, 25 buses are powered by natural gas, and the remaining 244 are electric trolleys (Acres, 1998). The specific breakdown of natural gas/diesel/trolley buses measured was not recorded, but it is common for buses of both types to travel on the same routes. Accordingly, bus travel may reflect travel on a street of higher (diesel) traffic density rather than exposures specifically due to the bus on which the subject was riding. Diesel buses were identified as a major source of direct PM2.5 emissions in a previous study (Balogh et al., 1994) and have been associated with high particle counts measured inside a moving automobile (Alm et al., 1997).

Cyclists in the greater Vancouver area travel on public roadways. For this reason, we expected particulate exposures while cycling to exceed many of the other modes of transportation. This turned out not to be the case. In fact, exposures while cycling were lower than all other modes of transportation for size fractions smaller than 5 µm. One possible explanation is that cyclists tend to travel during periods of lower traffic density. The cycle routes followed during this study were all high volume commercial roads. A study of Amsterdam cyclists' exposure to gaseous air pollutants also indicated that their exposures were lower than those encountered by car drivers on the same route, suggesting that particle exposures might also follow the same pattern (Van Wijnen et al., 1995). Elevated exposures to particles larger than 5 μm while cycling may be due to resuspended road dust associated with the bicycle itself or with surrounding vehicles. It is likely that a high percentage of resuspended coarse particles would be removed in the air handling system of cars and buses and therefore exposures to these size particles would be lower so long as windows were closed. The cycling exposures to particulate larger than 5 μm were significantly higher than those experienced while walking or commuting by car.

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Our measurements also demonstrated the important impact of ETS and cooking on fine particle exposures. This finding has also been clearly demonstrated in the PTEAM study in which elevated indoor PM_{2.5} and PM₁₀ levels were associated with cooking and ETS (Ozkaynak et al., 1996). We also found that even outdoor exposures to ETS were relatively common and resulted in elevated particle exposures (Figures 4–5). As demonstrated in numerous studies using gravimetric fine particle measurements (reviewed in Wallace, 1996), outdoor particle levels were higher then indoor levels, provided that no indoor particle sources were operating. In our measurements, we found that, on average, indoor particle counts were 62% of outdoor counts for particles <5 μm .

Conclusions

We found a low correlation between personal samples of particle counts and personal PM25 mass concentrations. However, despite this low correlation, we found the handheld particle counter, combined with a PDA, to be an effective method for identifying short-term particle concentrations peaks. This approach allowed us to assign measured particle exposures which lasted for only several minutes or less to specific activities and/or microenvironments. The PDA and associated software were simple, easy to use and provided electronic time-activity data with a high level of temporal resolution which was directly transferred into a computer for data analysis. Stratification of the measured particle counts by the different particle sources enabled us to identify activities/microenvironments which were associated with higher exposures. Outdoor particle counts were significantly higher than indoor counts for particles smaller than 5.0 μ m. Elevated fine particle exposures were associated with commuting by vehicles powered with internal combustion engines (bus, car, and Seabus), and with proximity to ETS, cooking, and wood smoke

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